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Efficiencies of freshwater and estuarine constructed wetlands for phenolic endocrine disruptor removal in Taiwan

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ABSTRACT

We examined the distribution and removal efficiencies of phenolic endocrine disruptors (EDs), namely nonylphenol diethoxylates (NP_2EO), nonylphenol monoethoxylates (NP_1EO), nonylphenol (NP), and octylphenol (OP), in wastewater treated by estuarine and freshwater constructed wetland systems in Dapeng Bay National Scenic Area (DBNSA) and along the Dahan River in Taiwan. Water samples were taken bimonthly at 30 sites in three estuarine constructed wetlands (Datan, Pengcun and Linbian right bank (A and B)) in DBNSA, for eight sampling campaigns. The average removal efficiencies were in the range of 3.13–97.3% for wetlands in DBNSA. The highest average removal occurred in the east inlet to the outlet of the Tatan wetland. The most frequently detected compound was OP (57.7%), whose concentration was up to 1458.7 ng/L in DBNSA. NP was seen in only 20.5% of the samples. The temporal variation of EDs showed a decrease across seasons, where summer > spring >winter > autumn in these constructed wetlands. The removal efficiencies of EDs by estuarine wetlands, in decreasing order, were Datan > Pengcun > Linbian right bank in DBNSA. Water samples collected at 18 sites in three freshwater constructed wetlands (Daniaopi, Hsin-Hai I, and Hsin-Hai II) along the riparian area of Dahan River. NP_2EO was the most abundant compound, with a concentration of up to 11200 ng/L. Removal efficiencies ranged from 55% to 91% for NP_1EO, NP_2EO, and NP in Hsin-Hai I. The average removal potential of EDs in freshwater constructed wetlands, in decreasing order, was Hsin-Hai II > Daniaopi > Hsin-Hai I constructed wetlands. The lowest concentrations of the selected compounds were observed in the winter. The highest removal efficiency of the selected phenolic endocrine disruptors was achieved by Hsin-Hai I wetland. The calculated risk quotients used to evaluate the ecological risk were up to 30 times higher in the freshwater wetlands along Dahan River than in the estuarine (DBNSA) constructed wetlands, indicating that existing concentrations of these EDs in wetland systems pose a high ecological risk to aquatic organisms. The decreasing risk quotient from influent to effluent indicates that phenolic endocrine disruptors can be treated in these constructed wetlands. Our results of this research can serve as a preliminary understanding on the ED removal efficiencies in different types of constructed wetlands.

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1. Introduction

Alkylphenol polyethoxylates (APEs), a class of nonionic surfactants, have been widely used in industrial, agricultural, and household applications. These compounds are added to emulsifiers, adhesives, and pesticides and tend to be adsorbed by sediment (Ying et al., 2002). APEs are lipophilic, recalcitrant in the environment, and easily absorbed by organisms; therefore, they have potential for bioaccumulation. Moreover, the metabolites of biodegradation of APEs such as alkylphenol, shortened ethoxy chain APE residues, alkylphenol polyethoxy carboxylates, and carboxyalkyl phenoxy ethoxycarboxylates, are more persistent than the parent APEs and are known to disrupt endocrine function in biota (Vethaak et al., 2005; Ce'spedes et al., 2005; Wang et al., 2006; Li et al., 2008). In Taiwan, nonylphenol polyethoxylated (NPEO) compounds are the most widely used nonionic surfactants (80%), followed by octylphenol ethoxylated (OPEO) compounds (20%) (Wang et al., 2001). The degradation metabolites of NPEOs and OPEOs such as nonylphenol (NP) and octylphenol (OP), are more toxic than the parent compound and are known to disrupt endocrine function in wildlife and humans.

It has been reported that the acute toxicity level (LC_{50}) of NP in freshwater fish (e.g., *Pimephales promelas* was 0.007–0.128 mg/L (Brooke, 1993; Ward and Boeri, 1991), and the LC_{50} for marine fish (e.g., *Pleuronectes americanus* and *Cyprinodon variegatus*) was 0.017–0.21 mg/L (Ward and Boeri, 1990). Acute toxicity levels for aquatic invertebrates (e.g., *Hyalella azteca* and *Ceriodaphnia dubia*) and midge (*Chironomus tentans*) were reported as 0.02–3 mg/L (Brooke, 1993; England, 1995; England and Bussard, 1995). Both NP and OP can induce vitellogenin (VTG) synthesis and feminization in male fish at the ppb concentration level, and the estrogenic activity of OP is 20 times that of NP according to previous research (Servos, 1999). Of critical

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importance, these compounds can enter humans via cutaneous absorption, ingestion, or inhalation, and affect endocrine function, especially in newborns and fetuses.

For ubiquitous contaminants, technology for treating wastewater includes wastewater treatment plants (WWTPs) and constructed wetlands. Many investigators have reported that pollutants such as nutrients and pathogens in wastewater can be removed efficiently using these systems to meet water quality criteria (Tsihrintzis et al., 2007; Juang and Chen, 2007; Galbrand et al., 2008; Lin et al., 2008). The removal efficiency of wetlands and WWTPs for a wide range of xenobiotic compounds is a topic of much current interest because the pollutant loads discharge into the environment can be significantly decreased. Several studies have demonstrated that there is limited efficiency in traditional WWTP processes for treating endocrine-disrupting chemicals (Gonza'lez et al., 2007). According to research, 40%–45% of persistent organic pollutants remain in the sewage treatment plant, and residues discharge to nearby rivers and accumulate in aquatic organisms through the food chain (Ahel et al., 1994; Clara et al., 2007; Stasinakis et al., 2008).

The distribution and removal of endocrine disruptors (EDs) such as alkylphenolic compounds in constructed wetland systems have been limited. A variety of small-scale domestic sewage treatment systems have been developed to remove compounds originating in pharmaceutical and personal care products. Vertical-flow constructed wetlands were found to be more common and efficient among different types of wetlands regarding to the higher removal efficiency (Matamoros et al., 2008). Conkle et al. (2008) found >90% removal of pharmaceutically active compounds (PhACs) in a lagoon wetland, similar to that obtained in conventional activated-sludge WWTPs. Song et al. (2008) developed a vertical-subsurface-flow constructed wetland mesocosm to remove estrogens, and obtained removal efficiencies >72% with a 7.5-cm plant rooting depth.

The present study determines the spatial and temporal variations of four phenolic endocrine disruptors, nonylphenol diethoxylate (NP_2EO), nonylphenol monoethoxylate (NP_1EO), nonylphenol (NP), and octylphenol (OP), in freshwater and estuarine constructed wetland systems located in the north and south of Taiwan. Removal efficiencies are assessed and the potential ecotoxicological risks of these compounds are evaluated in the constructed wetlands to understand potential hazards posed to aquatic organisms.

2. Material and methods

2.1. Site description

Water samples were collected from six constructed wetlands of two major wetland systems, Dapeng Bay National Scenic Area (DBNSA) and nearby Dahan River, located in the south and north of Taiwan, respectively. Three estuarine wetlands were sampled in DBNSA: Datan, Pengcun, and Linbiain; and three freshwater wetlands along the Dahan River: Daniaopi, and Hsin-Hai Bridge Phases I and II (Fig. 1).

2.1.1. Constructed wetlands along Dahan River

Samples were collected from 18 sites in the Daniaopi, and Hsin-Hai I and II constructed wetlands along the riparian area of Dahan River (Fig. 1A), Daniaopi constructed wetland is designed to treat 11000 m^3/day of wastewater. The eight sampling sites were located in the depositional pond (DN1), open water surface zone (DN2), fully vegetated zone I (DN3-1, DN3-2, DN3-3, DN3-4), fully vegetated zone II (DN4), and the ecological pond (DN5). Five sampling sites were located in each of Hsin-Hai I and II constructed wetlands, designated treatment units including a depositional pond (HS1-1 and HS2-1), fully vegetated zone I (HS1-2 and HS2-2), open water zone (HS1-3 and HS2-3), fully vegetated zone II (HS1-4 and HS2-4), and ecological pond (HS1-5 and HS2-5). The design flow rates for Hsin-Hai I and II constructed wetlands were 2200 and 3000–4000 m^3/d, respectively. Influent consists primarily of domestic wastewater.

2.1.2. Constructed wetlands in DBNSA

In DBNSA, a total of 30 sampling sites were selected in Datan, Pengcun, and Linbian right bank constructed wetlands (Fig. 1B). The Datan CW covers 6 ha and has design capacities of 3326 and 2635 m^3/d for the east and west side influents respectively. The nine sampling sites were designated as west influent (TW1), west depositional pond (TW2), west subsurface pool (TW3), east influent (TE1), east depositional pond (TE2), east side subsurface filtering basin (TE3), convergence site in marsh area (T4), ecological pond (T5), and effluent (T6). Pengcun CW occupies 7.4 ha with a design capacity of 9104 m^3/d. The eight selected sampling sites were designated as inflow in depositional pond (B1), filtering basin (B2), graved filter bed (B3), shallow water pond #1 (B4), shallow water pond #2 (B6), deep water ponds #3 and #6 (B3 and B7), and effluent (B8). The Linbian right bank CW receives influent from two sources (domestic and aquaculture wastewater). The design capacity was 9861 m^3/d for area A (a mix of domestic: 5961 m^3/d and aquaculture: 3900 m^3/d wastewater) and 6967 m^3/d for area B, which receives a combination of wastewater inflows. The selected sampling sites were located at the outlet of each unit. The configuration of the seven sampling sites in area A of Linbian CW was as follows: influent (LA1), depositional pond (LA2), subsurface filtering pond (LA3), influent and middle sections of marsh (LA4 and LA5), shallow marsh (LA6), and effluent (LA7). In area B, the eight selected sampling sites included influent (LB1), depositional pond (LB2), subsurface and filtering basin (LB3), effluent of subsurface and filtering basin (LB4), deep-water pond (LB5), middle and post sections of the marsh pond (LB6, LB7), and effluent (LB8).

2.2. Sampling

Grab samples were collected from December 2008 to January 2010 (eight campaigns) in the estuarine (DBNSA) constructed wetland system. Samples were not collected from area A in Linbian right bank constructed wetland due to the impact of Typhoon Morakot in September 2009. In addition, in December 2008 samples were collected from the settling basin and outlet site at the initial stage only, and these data were used for reference only. At Dahan River, along the riparian area, samples were collected quarterly from the freshwater constructed wetlands from November 2008 to December 2009. We followed the procedures presented in the final report of previous study, the pH of samples were adjusted to less than 2 with an acid to inhibit biological degradation until further analysis (Drewes et al., 2006).

2.3. Sample pretreatment and analysis

Three liters of each water sample was acidified and filtered through a 1 μm glass fiber filter and a 0.45 μm acetate cellulose filter to remove particles. Samples were then concentrated by solid-phase extraction followed by cartridge activation. Cartridges (OASIS HLB, 6 mL, 200 g) were used, and activated in the sequence of 10 mL dichloromethane/methanol (1/1, v/v), 5 mL methanol, and 10 mL deionized water. Water samples passed through the cartridge at a rate of 4 mL/min and were then rinsed with 10 mL double-distilled H_2O and 400 μL methanol, and 7 mL methylene chloride/methanol (1/1, v/v). The elutes were collected and blown nearly dry under nitrogen at a constant temperature of 40 °C. The samples were then concentrated with 2 mL of H_2O/acetonitrile and the final solution was filtered through a 0.45 μm polyvinylidene difluoride (PVDF) membrane filters before analysis. Chromatographic separation was achieved by liquid chromatography (Waters, USA) with a fluorescence detector (Agilent, USA) with XTerra column(5 μm, 4.6 × 250 mm) supplied by Waters (USA). This allowed us to detect all four analytes using a single pair of wavelengths ($\lambda_{em}/\lambda_{ex}$ 313/227 nm) with a reaction time of 16 min

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